

Universal observation of multiple order parameters in cuprate superconductors

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The temperature dependence of the London penetration depth λ was measured for an untwined single crystal of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ along the three principal crystallographic directions (a , b , and c). Both in-plane components (λ_a and λ_b) show an inflection point in their temperature dependence which is absent in the component along the c direction (λ_c). The data provide convincing evidence that the in-plane superconducting order parameter is a mixture of $s + d$ -wave symmetry whereas it is exclusively s -wave along the c direction. In conjunction with previous results it is concluded that coupled $s + d$ -order parameters are universal and intrinsic to cuprate superconductors.

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It is believed that the CuO_2 planes are the essential building units in cuprate high-temperature superconductors (HTS's) where superconductivity occurs. Even though either static or dynamic distortions of these planes destroy the cubic symmetry, many theoretical approaches ignore the observed orthorhombicity and idealize the planar structure, mostly in order to justify a pure d -wave order parameter. Early on it was, however, emphasized that cuprates must have a more complex order parameter than just d -wave [1, 2], supported by many experiments like nuclear magnetic resonance (NMR) [3], Raman scattering [4, 5], angle-resolved electron tunneling [6], Andreev reflection [7], angular-resolved photoemission (ARPES) [8], muon-spin rotation (μSR) [9, 10, 11], and neutron crystal-field spectroscopy [12]. In addition, experiments along the c axis like, *e.g.*, tunneling [13], bi-crystal twist Josephson junctions [14], optical pulsed probe [15], and optical reflectivity [16] suggest that a pure s -wave order parameter is realized here.

Multiple order parameter scenarios were proposed shortly after the BCS theory in order to account for a complex band structure and interband scattering [17, 18, 19]. This approach has the advantage that high-temperature superconductivity can easily be realized even within weak coupling theories since interband scattering provides a pairwise exchange between different bands which strongly enhances the transition temperature (T_c) as compared to a single band model. The first realization of two-band superconductivity has been made in Nb doped SrTiO_3 [20] and has not attracted very much attention. With the discovery of high-temperature superconductivity in MgB_2 two-gap superconductivity became more prominent, and meanwhile many more systems exhibiting multi-band superconductivity have been discovered (see, *e.g.*, Refs. [20, 21, 22, 23, 24]). Interestingly, in all these systems the coupled superconducting order parameters are of the same symmetry, *i.e.*, $s + s$, $d + d$.

In this respect HTS's are novel since here mixed order parameter symmetries are realized, namely, $s + d$. Theoretically it has been shown that mixed order parameters support even high values of T_c and lead to an almost doubling of the transition temperature as compared to coupled order parameters of the same symmetry [25].

In order to prove that complex order parameters are intrinsic and universal to HTS, previous μSR measurements [9, 10, 11] were continued for another HTS family, namely $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. The μSR technique has the advantage that it is bulk sensitive and a direct probe of the London penetration depth which is highly anisotropic in HTS's. Recent results for $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$ and $\text{YBa}_2\text{Cu}_4\text{O}_8$ [9, 10, 11] clearly demonstrate the existence of two coupled $s + d$ -wave gaps in the CuO_2 planes and an s -wave gap along the c axis in $\text{YBa}_2\text{Cu}_4\text{O}_8$. While these findings already suggest that a complex gap structure is intrinsic to HTS, the new results on $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$, presented below, support this conclusion consistently. We are thus reasoning that $s + d$ -wave superconductivity in the planes and s -wave superconductivity along the c direction are intrinsic and universal to this complex material class. In addition, this finding imposes serious constraints for theoretical models, *e.g.*, neither purely electronic nor 2D based approaches capture this complicated picture.

The crystal was grown by a crystal pulling technique [26] and exhibited a rectangular shape of approximate size of $4 \times 4 \times 1 \text{ mm}^3$. The sample was detwinned by annealing it under stress for 2 months at 400°C . The total fraction, where a and b axis are exchanged, occupies approximately 8 to 10% of the entire crystal, as confirmed by measurements with a polarized microscope. T_c and the transition width were determined by DC-magnetization measurements and found to be 91.2 K and 2 K, respectively.

The transverse-field μSR experiments were carried out

at the π M3 beam line at the Paul Scherrer Institute (Villigen, Switzerland). The samples were field cooled from above T_c to 1.7 K in magnetic fields ranging from 0.012 T to 0.64 T. The typical counting statistics were ~ 20 -25 million muon detections per experimental point. The experimental data were analyzed within the same scheme as described in Refs. 9, 10, 11. This is based on a four component Gaussian fit of the μ SR time spectra where one component describes the background signal stemming from muons stopped outside the sample, and the three other components describe the asymmetric local magnetic field $P(B)$ distribution in the superconductor in the mixed state. The magnetic field penetration depth λ was derived from the second moment of $P(B)$ since $\lambda^{-4} \propto \langle \Delta B^2 \rangle \propto \sigma_{sc}^2$ [27]. The superconducting part of the square root of the second moment ($\sigma_{sc} \propto \lambda^{-2}$) was obtained by subtracting the normal state nuclear moment contribution (σ_{nm}) from the measured σ , as $\sigma_{sc}^2 = \sigma^2 - \sigma_{nm}^2$ (see Ref. 9 for details).

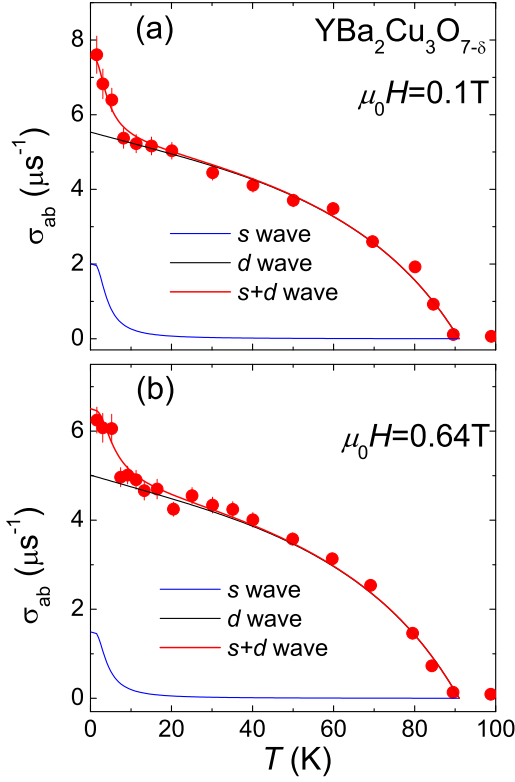


FIG. 1: (Color online) Temperature dependences of $\sigma_{ab} \propto \lambda_{ab}^{-2}$ of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ measured after field cooling in $\mu_0 H = 0.1$ T (a) and 0.64 T (b). The red lines represent results of a numerical calculation using the two-gap model [9, 10, 11] with parameters as summarized in Table I. The contributions of the small s -wave and the large d -wave gap to the in-plane superfluid density are shown by the blue and the black lines, respectively.

Since cuprates are highly anisotropic, the relation $\sigma_{sc} \propto \lambda^{-2}$ has to be extended to account for magnetic

fields applied along the three crystallographic directions ($i, j, k = a, b, c$). For the field applied along the i -th principal axis the penetration depth is determined from the second moment like $\lambda_{jk}^{-2} = (\lambda_j \lambda_k)^{-1} \propto \sigma_{jk} = \sqrt{\sigma_j \sigma_k}$ [28]. Here the index sc was omitted for clarity. The magnetic field dependence of the in-plane penetration depth has first been measured for different fields (0.05 T, 0.1 T, 0.2 T, and 0.64 T). The field was applied along the crystallographic c axis and, subsequently, the sample was cooled down from above T_c to 1.7 K. For this field configuration the in-plane component of the superfluid density $\sigma_{ab} \propto \lambda_{ab}^{-2} = (\lambda_a \lambda_b)^{-1}$ was obtained. In Fig. 1 σ_{ab} is shown as a function of temperature for two representative fields of 0.1 T and 0.64 T. In analogy to previous results [9, 10, 11], an inflection point in $\sigma_{ab}(T)$ is observed at $T \simeq 10$ K, which is a typical signature for the coexistence of a small s -wave and a large d -wave gap. Accordingly, the analysis of the data was performed by assuming that $\sigma(T)$ can be decomposed into two components having d -wave and s -wave symmetry as $\sigma(T) = \sigma^s(T) + \sigma^d(T)$ [9, 10, 11]. The temperature dependences of the individual components were obtained within the same framework as presented in Refs. 9, 10, 11. The comparison of experimental and theoretical results is made in Fig. 1, where the red lines refer to the sum of the two components, whereas the blue and the black lines display the individual s - and d -wave contributions, respectively. For all magnetic fields the analysis was based on common zero-temperature gap values (Δ_0^s and Δ_0^d) but field dependent second moments [$\sigma^s(0)$, $\sigma^d(0)$]. The zero-temperature gap values obtained in this way are $\Delta_0^s = 0.707(11)$ meV, $\Delta_0^d = 22.92(9)$ meV, and are in good agreement with results from tunneling experiments for the d -wave gap (see, *e.g.*, Ref. 29). The parameters obtained from the analysis are summarized in Table I.

The d -wave contribution to the total superfluid density $\omega = \sigma^d(0)/[\sigma^s(0) + \sigma^d(0)]$ increases with increasing field (see Fig. 2), similar to the field dependence observed for $\text{La}_{1.83}\text{Sr}_{0.17}\text{CuO}_4$ [9]. This dependence can be understood by the fact that superconductivity is suppressed stronger in the s -wave band with increasing field than in the d -wave band [9].

The individual components of the inverse squared penetration depth (λ_a^{-2} , λ_b^{-2} , and λ_c^{-2}) as functions of temperature were obtained by applying the magnetic field along the three crystallographic axes (0.012 T along a and b , and 0.1 T along c). This yields the axis-related superfluid densities according to [11]:

$$\sigma_i = \sigma_{ij} \cdot \sigma_{ik} / \sigma_{jk} \propto \lambda_i^{-2}. \quad (1)$$

The results are shown in Fig. 3 (a). Obviously σ_a and σ_b have a very similar temperature dependence, in particular, the inflection point at $T \simeq 10$ K and the linear increase in an intermediate range of temperatures ($60 \text{ K} \gtrsim T \gtrsim 10 \text{ K}$). The temperature dependence of

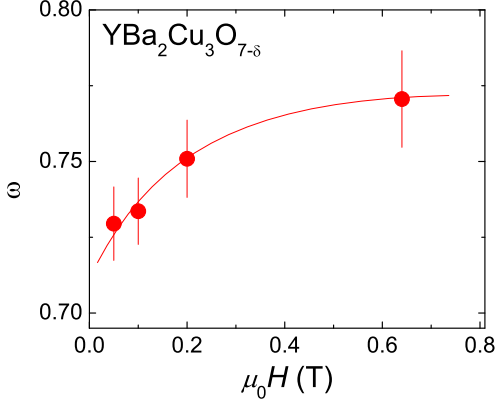


FIG. 2: (Color online) The d -wave contribution to the in-plane superfluid density $\omega = \sigma^d(0)/[\sigma^s(0) + \sigma^d(0)]$ as a function of the magnetic field of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. The line is a guide to the eye.

TABLE I: Summary of the two-gap analysis for untwined single-crystal $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ for the magnetic field applied along the c direction. The meaning of the parameters is – $\mu_0 H$: external magnetic field, $\sigma^d(0)$ and $\sigma^s(0)$: d -wave and s -wave contribution to the zero-temperature μSR relaxation rate $\sigma(0)$, $\omega = \sigma^d(0)/[\sigma^s(0) + \sigma^d(0)]$: the contribution of the large d -wave gap to the total in-plane superfluid density, Δ_0^d : d -wave gap at $T = 0$ K, Δ_0^s : s -wave gap at $T = 0$ K.

$\mu_0 H$ (T)	$\sigma^d(0)$ (μs^{-1})	$\sigma^s(0)$ (μs^{-1})	ω	Δ_0^d (meV)	Δ_0^s (meV)
0.05	1.78(2)	4.80(7)	0.729(12)		
0.1	2.01(2)	5.53(6)	0.734(11)		
0.2	1.87(2)	5.63(7)	0.751(13)	22.92(9)	0.707(11)
0.64	1.49(2)	5.01(7)	0.771(16)		

σ_c is qualitatively very different from the one of σ_a and σ_b . Here a saturation is observed at $T \lesssim 40$ K. $\sigma_a(T)$ and $\sigma_b(T)$ can be well described by the two-component approach mentioned above where the same zero-temperature gap values were used. From this analysis the following individual contributions from the s - and the d -wave components along the a and b axis are obtained: $\sigma_a^s(0) = 1.19 \mu\text{s}^{-1}$, $\sigma_a^d(0) = 3.83 \mu\text{s}^{-1}$ and $\sigma_b^s(0) = 2.51 \mu\text{s}^{-1}$, $\sigma_b^d(0) = 5.95 \mu\text{s}^{-1}$. Since the relative contributions of the large d -wave component are almost the same along a and b directions, namely, $\omega_a = 0.70$, $\omega_b = 0.76$, it is plausible to conclude that not the CuO chains are the cause of the two-component behavior [30], but that this is an intrinsic property of cuprates. The same conclusions were reached from different experimental techniques as, e.g., NMR [3], Raman scattering [4, 5], and ARPES [8]. In particular, Masui *et al.* [4] showed that the $s+d$ symmetry of the order parameter is required

in order to describe the Raman data, even for *tetragonal* $\text{Ti}_2\text{Ba}_2\text{CuO}_{6+\delta}$ HTS's.

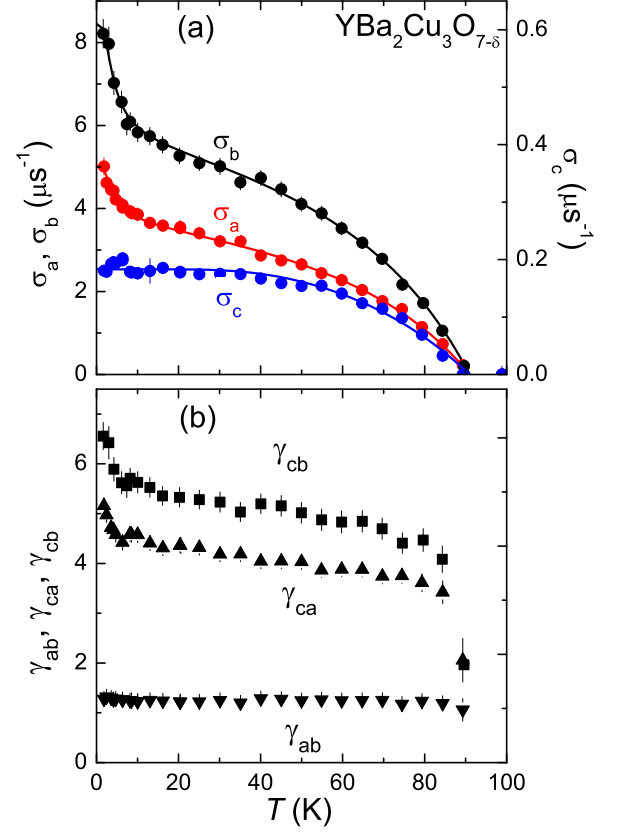


FIG. 3: (Color online) (a) Temperature dependences of $\sigma_a \propto \lambda_a^{-2}$, $\sigma_b \propto \lambda_b^{-2}$, and $\sigma_c \propto \lambda_c^{-2}$ of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ obtained from $\sigma(T)$ measured along the crystallographic a , b , and c directions by using Eq. (1). Lines represent results of the analysis within the two-component (black and red lines) and one-component (blue line) models [11]. (b) Temperature dependences of the anisotropy parameters γ_{ab} , γ_{ca} , and γ_{cb} obtained as $\gamma_{ij} = \lambda_i/\lambda_j = \sqrt{\sigma_j/\sigma_i}$ (see text for details).

The temperature dependence of the c axis related superfluid density $\sigma_{sc} \propto \lambda_c^{-2}$ resembles very much the one observed for $\text{YBa}_2\text{Cu}_4\text{O}_8$ [11] and follows closely the one expected for a single s -wave gap. The full blue curve in Fig. 3 (a) corresponds to an analysis based on an isotropic s -wave gap with $\Delta_0^s = 17.52$ meV and $\sigma_c(0) = 0.183 \mu\text{s}^{-1}$. The present results are in good agreement with previous findings of tunnelling experiments, where an s -wave gap along the c direction was reported [13]. The s -wave component along the c axis is not easily detectable by most experimental methods because either very well oriented films should be prepared or bulk methods have to be used. Due to the fact that many experiments are surface sensitive only, and ab oriented samples and films are hardly available, these techniques are unable to see the s -wave component along the c axis. Its observation is, however, important since

the coupling of a major d -wave component in the ab plane to the s -wave component along the c axis mixes both symmetries in the planes already. In addition, theory predicts for this scenario, that a pure d -wave order parameter is never observable. On the other hand, also along the c direction an admixture of the d -wave order parameter has to be present [25]. This latter statement could provide an explanation for the optical conductivity spectra along the c direction where strongly anisotropic gap like features have been observed together with a finite density of states at the Fermi energy [31, 32].

Finally, the anisotropy along all three crystallographic directions is addressed. This can be calculated by using Eq. (1) and defining the anisotropy as $\gamma_{ij} = \lambda_i/\lambda_j = \sqrt{\sigma_j/\sigma_i}$. The results are shown in Fig. 3 (b). While the in-plane anisotropy (γ_{ab}) is almost constant for all temperatures, both out-of-plane components (γ_{ca} and γ_{cb}) exhibit a sharp increase, as expected from Fig. 3 (a). In addition, it is seen that while γ_{ab} is almost always close to 1.2, γ_{ca} , γ_{cb} are substantially larger and reach values between 5 and 6.5 in the low- T limit. This high anisotropy is in very good agreement with previously reported values [33, 34]. It reflects the fact that the CuO_2 planes have nearly Fermi liquid-like metallic properties whereas along the c direction mostly insulating behavior is observed.

Our conclusions from the above presented data are manifold. Since $s + d$ -wave symmetries of the superconducting order parameter were observed previously in various cuprate families by various different techniques [3, 4, 5, 6, 7, 8, 9, 10, 11, 12], the new μSR data together with earlier results on structurally different compounds [9, 10, 11] support the idea that this behavior is *intrinsic* and *universal*. Similarly, the observation of an s -wave order parameter along the crystallographic c axis is proposed to be intrinsic as well. Specifically, this latter point emphasizes the importance of the *third* dimension for HTS's which was neglected in most of the theoretical models. In this context it is worth mentioning that the importance of the c axis has already been emphasized from ab-initio band structure calculations, where trends in T_c were correlated with CuO_2 apical oxygen distances [35]. Also, from first principles doping dependent computations of ARPES intensities, it was concluded that contributions from the c axis are of crucial importance in understanding the physics of HTS's [36]. On the other hand, the observation of mixed order parameters and more specifically, the additional s -wave component, require that the lattice must be considered in the physics of HTS's.

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